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CONTINUOUS-WAVE STIMULATED RAMAN EMISSION

SEMIANNUAL TECHNICAL SUMMARY REPORT

(For the Period Ending 31 October 1965)

Contract No. Nonr 4847(00)

ARPA Order No. 306

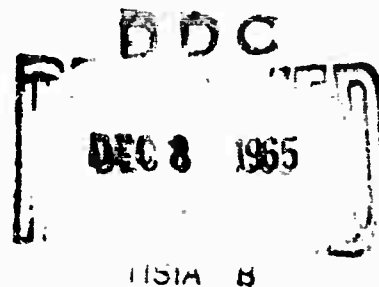
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Perkin-Elmer Engineering Report No. PE-TR-8186

Marvin C. Tobin
Project Scientist



The Perkin-Elmer Corporation
Electro-Optical Division
Norwalk, Connecticut 06852

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SUMMARY

A total of twenty-five organic liquids were examined in this measurement series. Stimulated Raman emission was observed in twenty-one organic liquids. In none of these was the strongest line near 1057 cm^{-1} , the frequency needed for continuous wave generation of stimulated Raman emission. In spontaneous Raman emission, however, three compounds were observed to have strong lines near 1057 cm^{-1} and will be used in the continuous wave experiment. Spontaneous line halfwidths were measured in some of these compounds.

A one-meter tank with glass walls was constructed and used with a Q-switch ruby laser to (1) observe light scattering in water and in organic liquids (2) look for effects ascribable to electrostriction and (3) measure a gain of 0.05 cm^{-1} for the 1004 cm^{-1} line of toluene in sample lengths of 50-100 cm.

The light scattered to the side showed discrete scattering sites to be present. No effects definitely ascribable to electrostriction could be observed.

SECTION I

INTRODUCTION

The goal of this program is the elucidation of discrepancies that occur between the theory of simulated Raman emission and experimental observations. In this program, attention is focused on discrepancies between stimulated Raman gains measured with pulsed excitation and gains calculated from spontaneous Raman scattering cross-sections and linewidths.

Major experiments involve illuminating a sample with two lines from a continuous-wave gas laser. It is intended to select samples that have a strong Raman shift at a wavenumber value corresponding to the wavenumber separation of the laser lines. With these at hand we will carry out the following:

- (1) Measurement of gain in the Stokes line, using a marginal oscillator to measure small changes in gain.
- (2) Measurement of stimulated anti-Stokes generation, by crossing the laser lines at the proper matching angle in the sample.

In pursuance of the main objective, we established some subsidiary objectives:

- (1) Measurement of spontaneous scattering cross-sections and linewidths for suitable lines in the samples selected for the main experiments.
- (2) Where necessary, measurement of stimulated Raman gains with a Q-switch ruby laser.
- (3) Search for effects ascribed to electrostriction, cavitation, etc., during stimulated Raman generation with a Q-switch ruby laser.

We decided to undertake the measurement of continuous-wave stimulated anti-Stokes emission as our first goal, selecting the 20487 and 19430 cm^{-1} argon laser lines, with a 1057 cm^{-1} separation as our working lines.

Our accomplishments to date are the following:

- (1) We have examined twenty-five compounds for stimulated Raman emission. Spontaneous line halfwidths have been measured in some of these.
- (2) We have completed preliminary experiments using an argon laser as an exciting source for spontaneous Raman emission. We intend to use this to measure line halfwidths and scattering cross-sections.

- (3) We have photographed the light scattered sideways from a pulsed laser beam in liquids, using infrared film, and have drawn some conclusions from the character of the scattered light.
- (4) We have carried out some experiments, in a one-meter-long Raman cell, to search for effects due to electrostriction.
- (5) Using a Q-switch laser, we have made some approximate measurements of stimulated Raman gain for the 1004 cm^{-1} line in toluene, obtaining a figure of 0.05 cm^{-1} .
- (6) We have built a Raman cell for carrying out the stimulated anti-Stokes generation experiment.

We are now at the point where we are ready to attempt the experiment on stimulated Stokes generation. The main effort during the next quarter will be directed toward this experiment and toward the construction of an argon-laser-driven spontaneous Raman spectrometer for the measurement of scattering cross-sections.

SECTION II

STIMULATED RAMAN SPECTRA AND SPONTANEOUS LINE HALFWIDTHS

The results of our experiments on stimulated Raman emission are shown in Table I. These were obtained using the experimental techniques described in the first quarterly report.

In no instance did a line near 1057 cm^{-1} appear in stimulated Raman emission, although this might have been hoped for in some of the substituted benzenes. On the basis that a compound which emitted stimulated Raman radiation in any line should be a strong Raman scatterer in other lines, the stimulated emission was used as a screening test.

The compounds which gave spontaneous Raman emission were measured on a Perkin-Elmer LR-1 Raman spectrometer to locate lines near 1057 cm^{-1} . Only 1-chloronaphthalene (1057 cm^{-1}), 1,3-dibromobenzene (1055 cm^{-1}), and 1,2-diethyl benzene (1053 cm^{-1}) proved to have strong lines close to 1057 cm^{-1} . Since these lines have from 10-30% the strength of the strongest lines in their spectra, these compounds are suitable candidates for experimentation.

In addition to line positions, some measurements on line half-widths were made. This was done by measuring the apparent halfwidth at a number of instrument slit settings. The resultant straight-line plots were extrapolated to zero instrument slitwidth. The values given in Table II are thought to be accurate to within 10-20%. *

* A note entitled "Stimulated Raman Emission Frequencies in Twenty-One Organic Liquids," by J.J. Barrett and M.C. Tobin, has been accepted for publication by the Journal of the Optical Society of America.

TABLE I
STIMULATED RAMAN EMISSION FROM ORGANIC LIQUIDS

	Measured frequency shift of stimulated Raman line cm ⁻¹	Literature ¹ value for correspond- ing spontaneous Raman frequency shift cm ⁻¹
cis 1,2 - dimethyl cyclohexane	2853 2921	2854 2927
cis, trans 1,3 - dimethyl cyclohexane	2844 2866 2926	2844 2870 2931
cis 1,4 - dimethyl cyclohexane	2876	2873
2 - ethyl naphthalene	1382	1381
1 - chloronaphthalene	1374	1368
1 - fluoro - 2 - chlorobenzene ⁺	1034 3082	1030 3084
benzaldehyde*	1001	1000
t-butyl benzene	1000 3065	1002 3064
1,2 - diethylbenzene	2934	
1,3 - dibromobenzene	992	990
pyridine	991 3058	991 3054
2 - octene	2918	2908, 2931
cis 2 - heptene	2915	2920
2,3 - dimethyl- 1,5 - hexadiene	2910	
1 - hexyne	2116 2915	2119 2908
tetrachloroethylene*	448	447
2 - chloro - 2 - methylbutane	2927	2931
1 - bromopropane	2935 2962	
2 - bromopropane	2920	
2 - nitropropane	2945	2948
1,1,2,2 - tetrachloroethane	2986	2985

+very weak, diffuse line

*1:1 molar mixture of tetrachloroethylene and benzaldehyde gave two Stokes orders of benzaldehyde only.

DID NOT EMIT

1 - hexene
propionaldehyde
Sn (CH₃)₄
SnI₄ in CS₂
trichloroethylene

TABLE II

SPONTANEOUS LINE HALFWIDTHS IN ORGANIC LIQUIDS

	<u>Line Position, cm⁻¹</u>	<u>Line Halfwidth, cm⁻¹</u>
2 - ethyl naphthalene	1382	11.1
1 - chloronaphthalene	1057	6 (estimated)
	1374	8.2
t - butyl benzene	1026	3.8
	1000	2.8
1,3 - dibromobenzene	992	3.3
	1055	4.2
tetrachloroethylene	448	7.8

It is noteworthy that the line halfwidths measured vary over a narrow range, the widest line being only four times as wide as the narrowest. The intensities of neighboring Raman lines in a given compound (comparison of neighboring lines compensates for instrument sensitivity to wavelength) might vary by as much as several hundred times. It is likely, that at least in aromatic compounds, line intensity will be the predominant factor in determining the efficiency of stimulated Raman emission.

This part of the project is continuing. The results reported are those obtained to date.

SECTION III

PRELIMINARY EXPERIMENTS ON USE OF AN ARGON LASER
AS A RAMAN SOURCE

We have made some preliminary measurements on spontaneous Raman emission from benzene, using available laboratory equipment to assemble a Raman spectrometer.

The argon laser was adjusted, with a prism in the cavity, to emit either the 19430 cm^{-1} or the 20487 cm^{-1} line. The laser beam was deflected with a 90-degree prism to make it perpendicular to the table top and was passed through the bottom of a standard ultraviolet absorption cell. The image of the beam in the sample was focused onto the slit of a Jarrell-Ash 82-000 Monochromator fitted with an IP-28 photocell. This arrangement was satisfactory for recording Raman spectra.

Notable features of this simple arrangement are:

- (1) Even with the single-pass monochromator and with only a single preliminary filtration of the liquid, the spectra can be read to within $20\text{-}30\text{ cm}^{-1}$ of the exciting line.
- (2) The stronger Stokes Raman lines are quite bright to the naked eye through the exit slit, even in a well-lighted room.

- (3) If the sample is viewed through a small Bausch and Lomb monochromator, the anti-Stokes line of 986 cm^{-1} is clearly visible to the naked eye in a darkened room

By comparison, using mercury arc excitation under similar conditions, the 986 cm^{-1} Stokes line is just barely visible to the dark-adapted eye.

We are in the process of establishing this equipment on a permanent basis to measure linewidths and cross-sections and to try the stimulated anti-Stokes experiment. A low-dark-current EMI phototube with an "S" surface has been obtained and will be calibrated with the Jarrell-Ash monochromator to read absolute light fluxes. We are also obtaining a double monochromator for the detection of very weak signals at high resolution. This will replace the Jarrell-Ash monochromator at a later date.

SECTION IV

LIGHT SCATTERING EXPERIMENTS

The purpose of this series of experiments was to look for visible evidence of damage to liquid samples illuminated with a focused Q-switch laser beam.

For this and the succeeding experiments, a one-meter tank with glass sides was built so that the laser beam could be viewed perpendicular to its path.

For other experiments, small boats with glass sides were built to ride in the tank. Finally, a photocell was fitted with an extension arm and an opal glass diffuser to ride in the tank. The laser beam struck the opal glass and was scattered into the phototube. This configuration was used for the gain experiments (Section VI). The equipment is shown in Figure 1.

The laser beam was focused into various liquids with a 30-cm-focal-length lens placed near the inlet window. The tank was photographed from the side using Polaroid 413 infrared film. The tracks made by the laser beam did not show up on Polaroid 410 film (speed 10,000), showing that the tracks were not self-luminous. However, a piece of glass placed in benzene near the focus of the lens was damaged, with a visible white spark showing.

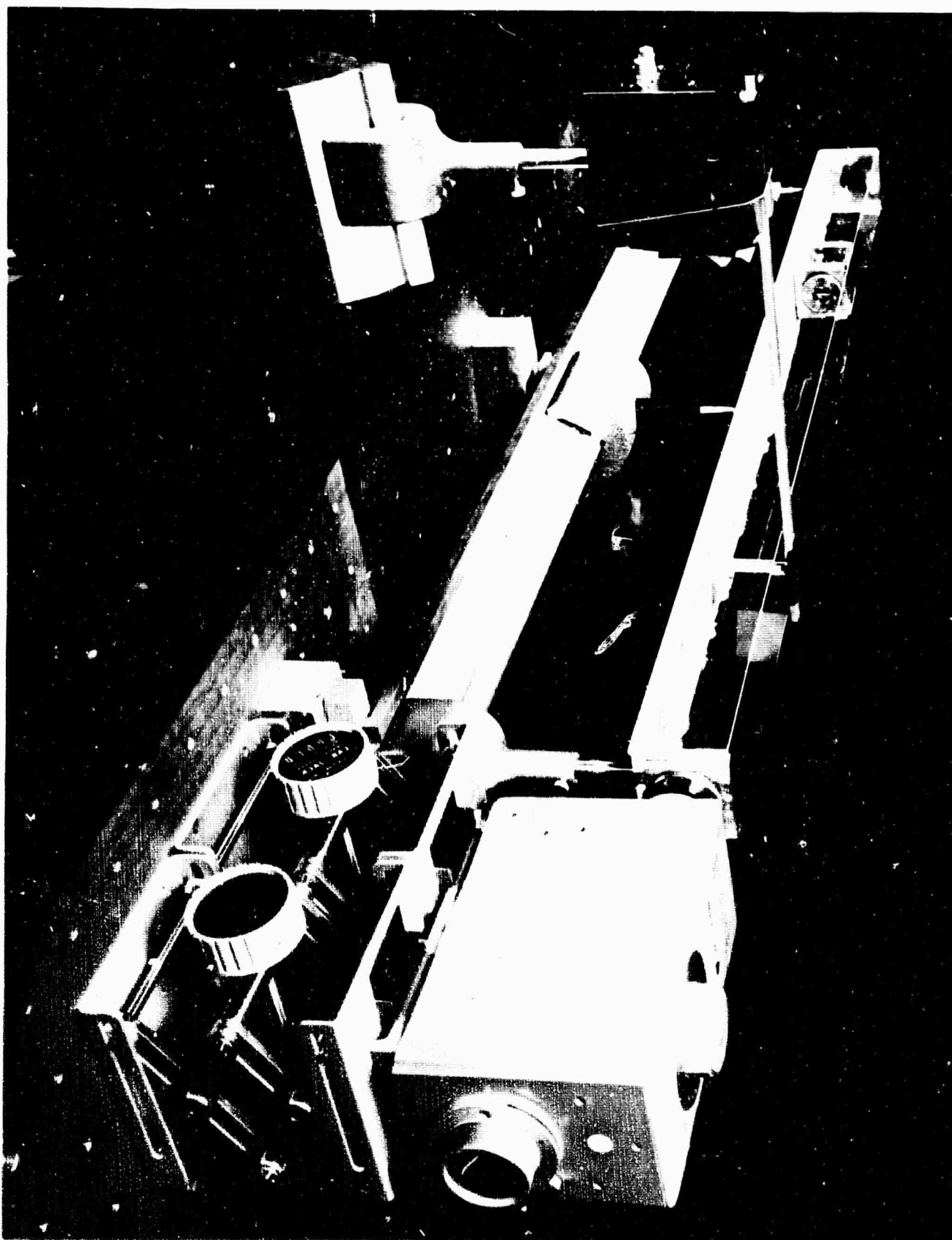


Figure 1. Glass Tank and Boats

The track in benzene could be photographed with Schott RG-9 plus RG-10 filters (sharp-cut long-pass opening near 7200A) before the camera lens, whereas the track in water could not. This showed that Stokes, as well as laser radiation was being scattered from benzene.

Figure 2 is a photograph of benzene in the tank and water in the 15-cm boat. Figure 3 shows a photograph with water in the tank and tetrachloroethylene in the 15-cm boat.

Of the materials photographed, (water, benzene, trichloroethylene and tetrachloroethylene) water was the only material in which the track was filamentary. In all instances, if the camera aperture was stopped down, the tracks in the organic liquids were seen to have a granular structure.

These experiments show that the plane-wave front assumed in theories of stimulated Raman emission is being perturbed by some kind of scattering center. These centers could be either dust motes or damage centers. At least in water, the filamentary track implies the latter. Damage bubbles could be formed by dielectric breakdown even at the 25 megawatt/cm² field in our unfocused beam (25 megawatt/cm² \approx 10⁵ V/cm, about the dielectric strength of average liquids). Supposing the gas-liquid interface to move at the speed of sound (about 5 x 10⁵ cm/sec in water) a bubble could grow to a diameter of 150 microns in the 30 nanoseccnds our pulse lasted.

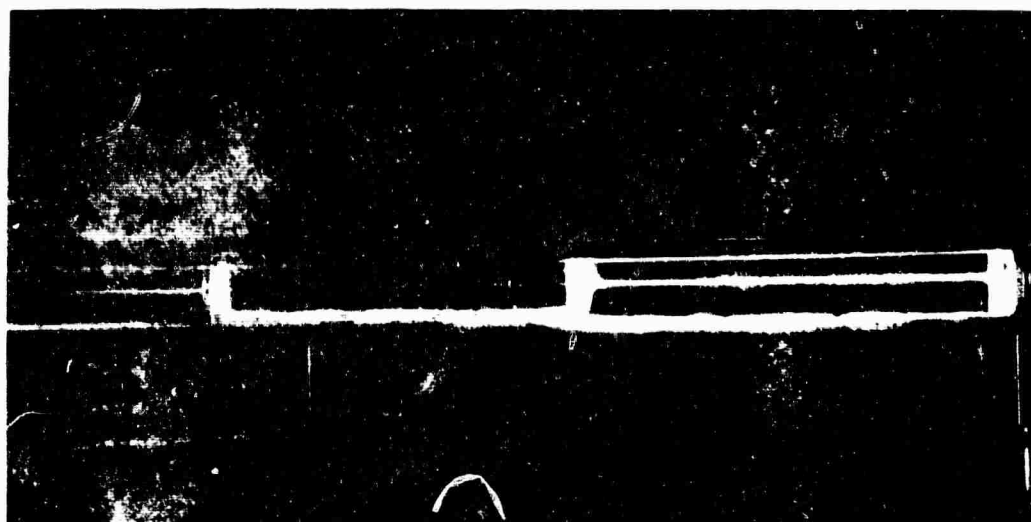


Figure 2. 30 cm Focal Length Focused Into 15 cm Boat Filled With Water in Tank Containing Benzene. Polaroid 413 Film.

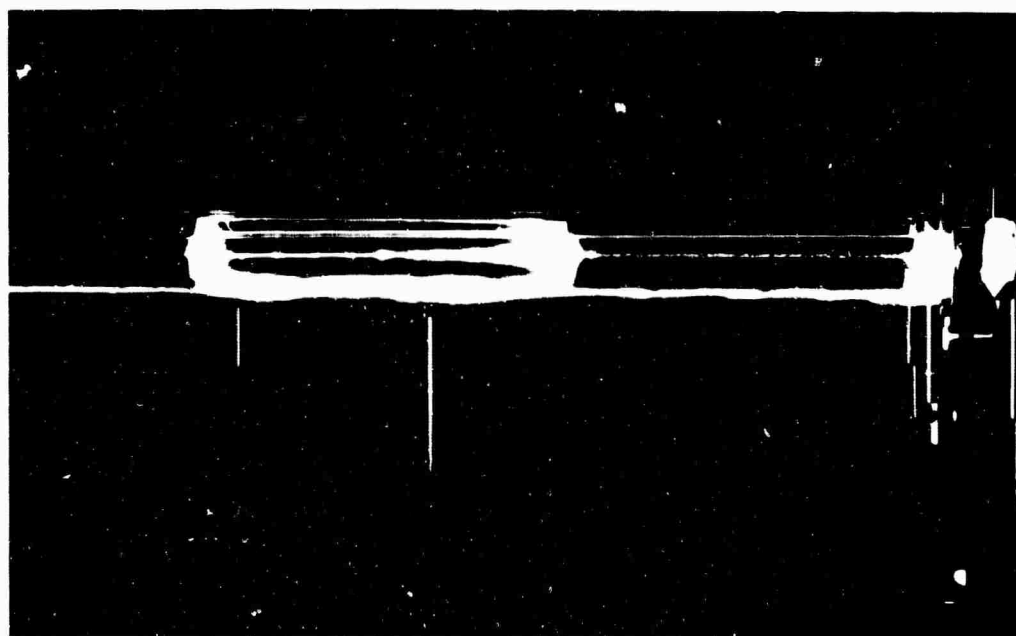


Figure 3. 30 cm Focal Length Focused Into 15 cm Boat Filled With Tetrachloroethylene in Tank Containing Water. Focal Region of Lens Inside Tetrachloroethylene. Polaroid 413 Film.

We do not intend to pursue these experiments further. Clearly, they should be tried with carefully filtered dust-free liquids, and with time-resolved photographs to see if the scattering centers are growing.

SECTION V

EXPERIMENTS TO SEARCH FOR ELECTROSTRICTION EFFECTS

This series of experiments was undertaken to determine whether any gross effects occurred in liquids which could be ascribed to electrostriction. In particular, it was desired to test whether or not any gross "downpuckering" of an unfocused laser beam occurred. In other words, attempts were made to discover if field-induced changes in the refractive index caused the liquid to act as a lens and focus the beam. If such an effect occurred, it could explain the reported anomalies in Raman gain data.

Three experiments were attempted:

1. Pieces of exposed Polaroid film were immersed in toluene, placed in different positions along the tank, and burned with an unfocused laser beam. The burn spot sizes were found to be independent of position along the tank and were similar to those obtained with the Polaroid outside the tank.
2. With the tank filled with 2-propanol ($n=1.38$), the 5-cm boat filled with benzene, and the opal glass in the boat, the scattered light was examined for Stokes radiation at various positions of the boat along the tank. No Stokes radiation was detected. An unfocused laser beam was used.
3. With the tank filled with water and the 5-cm boat filled with benzene, the laser beam was focused into the tank with a 30-cm focal length lens. The output out the far end of the tank was examined for Stokes

radiation. The only benzene Stokes radiation detected occurred when the boat was located near the calculated focus of the lens in the liquid.

Clearly, the toluene had no large-scale focusing effect on the unfocused laser beam. Similarly, neither 2-propanol with an unfocused beam, or water with a focused beam, had any gross effect on the process of Raman generation.

SECTION VI

MEASUREMENT OF STIMULATED RAMAN GAIN IN TOLUENE
USING A PULSED LASER

The equipment that was used to determine electrostrictive effects in liquids was also used to make approximate measurements of gain. These were made for the 1004 cm^{-1} line of toluene. Figure 4 is a schematic of the equipment layout.

A Tektronix 555 double-beam oscilloscope was used to monitor the laser beam entering the tank and the signal scattered from the opal glass. RCA 925 vacuum photo-diodes (S-1 surface) were used to detect the output. The RCA 925 vacuum photo-diodes showed a slight saturation at the light levels needed to generate a useful voltage across a 100Ω load resistor.

During the experiments, both the photocell viewing the laser pulse and the photocell viewing the opal glass were fitted with Corning 2-64 filters and white paper attenuators. The opal glass was placed inside the liquid five cm from the inlet window, and both oscilloscope channels were set at a gain of 0.005V/cm . The paper attenuators were then adjusted until both channels gave comparable signals. At this position of the opal glass, a combination of Corning 7-64 and Schott RG-9 and RG-10 filters (called here "triple filter, sharp-cutoff, long-pass at 7200\AA ") completely blocked any signal from reaching the phototube, showing the absence of Stokes radiation. At the same position of the opal glass, but without the triple filter, the signal was measured with Wratten 0.4, 0.5, and 1.0 neutral density filters before the phototube viewing

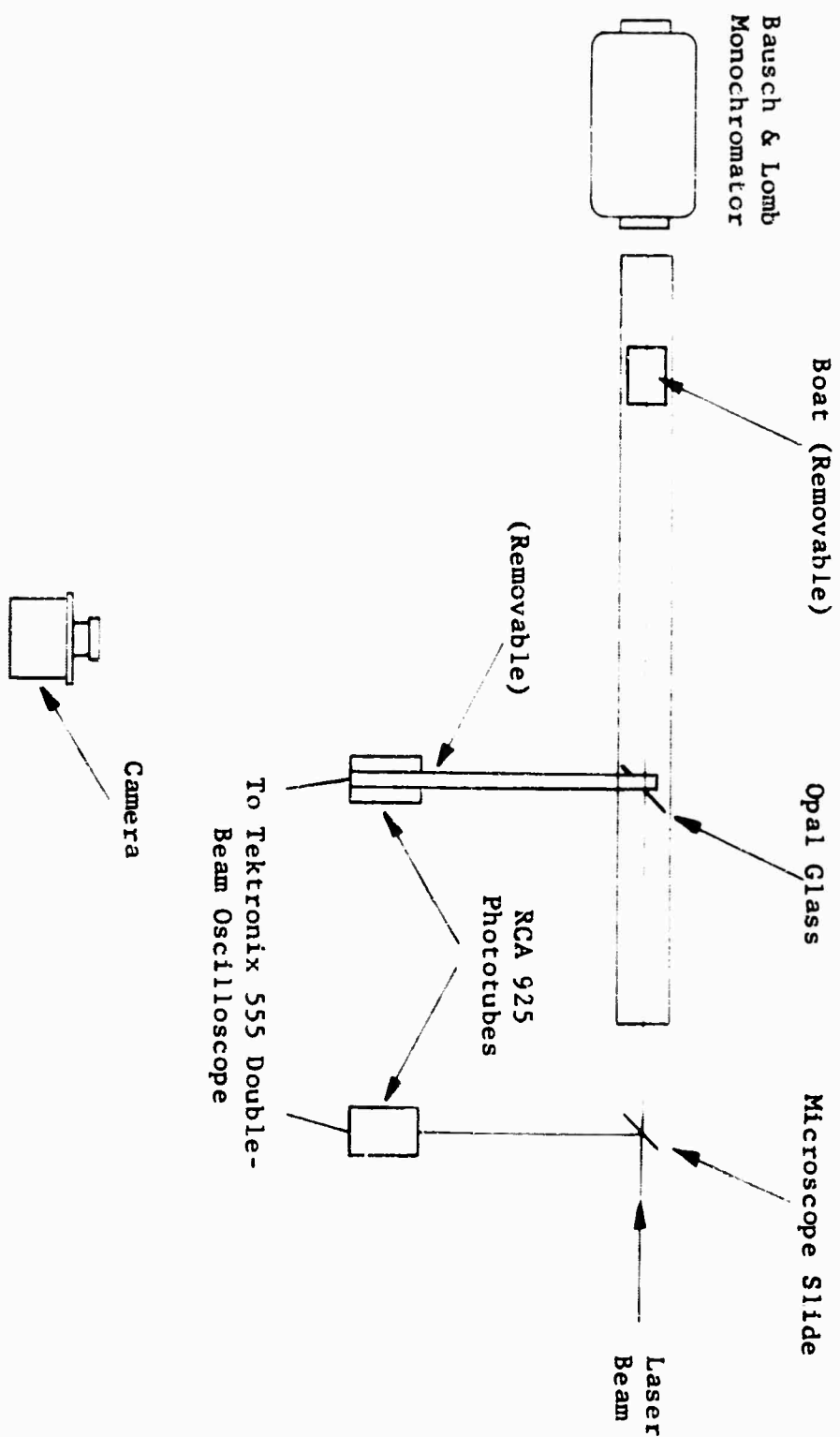


Figure 4. Setup Used in Gain Experiment, Light Scattering Experiment and Electrostriction Experiments

the opal glass. The data were plotted and used to correct for saturation of the phototubes or other nonlinearities of signal versus intensity. The opal glass was moved down the tank and, at each position of the glass, the signal was measured without the triple filter (laser & Stokes) and with it (part of Stokes).

The experiment was repeated on two successive days. On the third day, the opal glass scatterer was removed from the tank. A piece of opal glass was placed at the exit window of the tank. A small Bausch and Lomb monochromator set at 7470\AA was fitted with the phototube formerly used to view the opal glass, and was set to view the exit window of the tank. This arrangement was used to measure the transmission of the triple filter at 7470\AA . This turned out to be about 30%.

In a separate experiment, the energy content of the laser beam (0.22 ± 0.04 Joules) was measured with a TRG calorimeter and Keithley microvoltmeter. The area of a spot burned into a piece of Polaroid film at the position of the calorimeter was 0.31 cm^2 . These numbers yield a power density of 24 ± 4 megawatts/ cm^2 when combined with the 30 nanosecond pulse halfwidth measured on a Tektronix 519 oscilloscope.

The response of the S-1 surface of the RCA-925 phototube was corrected using the manufacturer's data for the S-1 surface. The response is, in fact, nearly flat in the region of interest.

As expected, the measured Stokes plus laser signal along the length of the tank was found to be nearly constant.

The raw data were corrected for phototube saturation, phototube sensitivity, and filter characteristics, and were plotted as percentage conversion

of input laser to Stokes radiation. The results of two independent experiments are shown in Figure 5. The experimental points (for liquid path lengths between 50 and 100 cm) fall on plots which are equivalent to:

$$P_{\text{Stokes}} \text{ (watts/cm}^2\text{)} = 5.8 \times 10^3 e^{-0.05L}$$

and

$$P_{\text{Stokes}} \text{ (watts/cm}^2\text{)} = 9.0 \times 10^3 e^{-0.05L}$$

In calculating gain in nitrobenzene, Weiner, Schwarz and McClung² measured the Stokes power exiting from a 10 cm long cell, assumed $P_0 \approx 10^{-6}$ watt/cm², and estimated a gain of about 3.0 cm⁻¹. In our experiment, we assumed no value for P_0 ; we calculated it and the gain from the data.

Thus, it appears that our long-cell experiments result in a gain which is theoretically reasonable, but yield an anomalous value for P_0 . The two sets of experiments can be reconciled if it is supposed that a high-gain regime appears in the first part of a long cell, producing a sizeable amount of Stokes radiation. Once this is present, the gain drops to the theoretical value.

We do not plan to pursue these experiments.

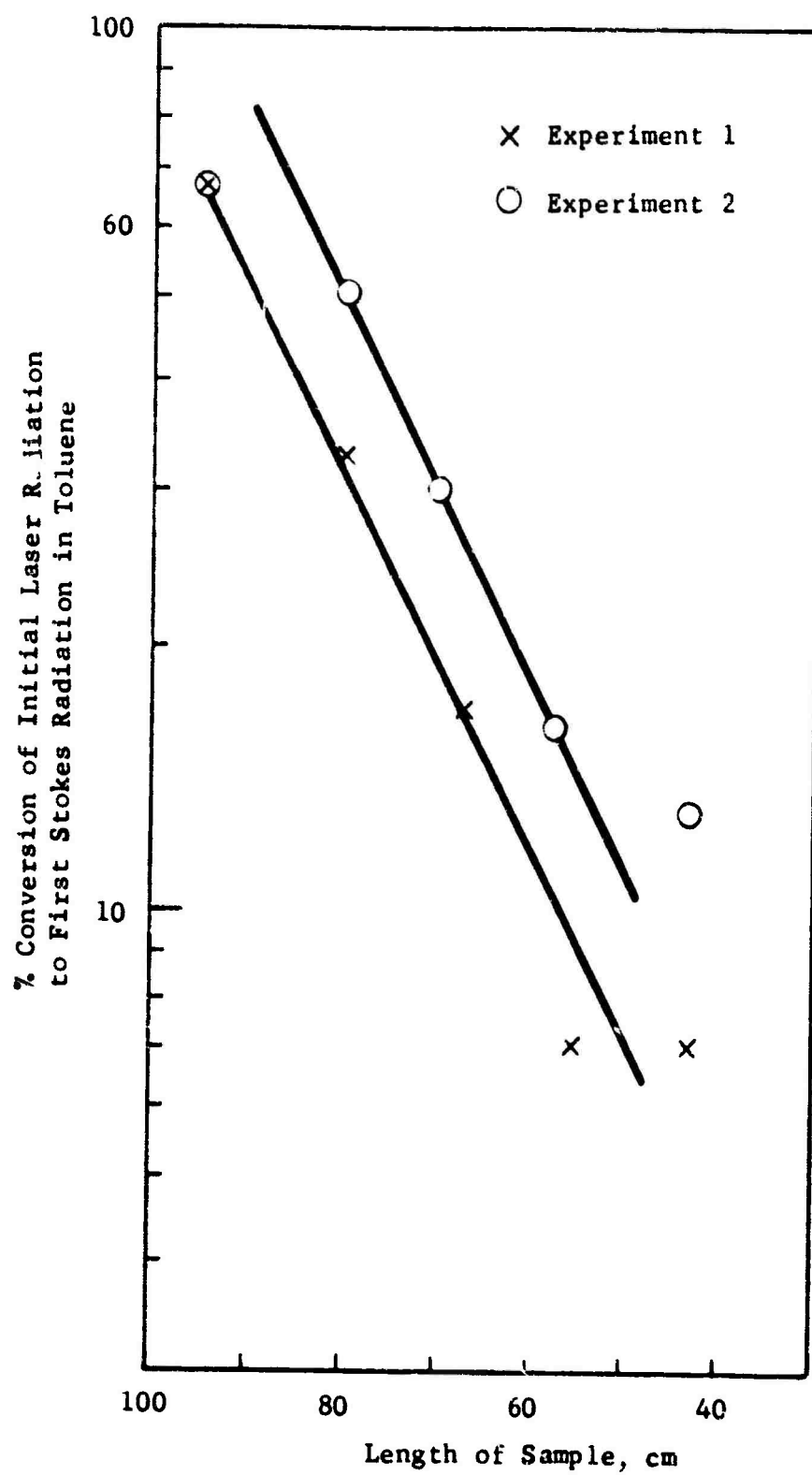


Figure 5.

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- ²D. Weiner, S. E. Schwartz and F. J. McClung, J. Applied Phys. 36, 2395(1965)

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Raman Emission
Stimulated
Spontaneous

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